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Signed Andrew Gersey

Dated 5 September 2000

Patents Form 1/77

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Request for grant of a patent

1.	Your Reference	**DPW/LJB/Q337**
2.	Application number	04 SEP 1999 **9920843.1**
3.	Full name, address and postcode of the or each Applicant Country/state of incorporation (if applicable)	**Sheffield Hallam University,** **City Campus,** **Howard Street,** **Sheffield.** **S1 1WB** 7444110003 **Incorporated in: The United Kingdom**
4.	Title of the invention	**A GLASS COMPOSITE**
5.	Name of agent Address for service in the UK to which all correspondence should be sent Patents ADP number	**APPLEYARD LEES** **15 CLARE ROAD** **HALIFAX** **HX1 2HY** **AA005** 190001
6.	Priority claimed to:	Country / Application number / Date of filing
7.	Divisional status claimed from:	Number of parent application: - / Date of filing: -
8.	Is a statement of inventorship and of right to grant a patent required in support of this application?	**YES**

A GLASS COMPOSITE

The present invention relates to a glass composite and, in particular, but not exclusively, a glass composite which utilises waste glass from consumer, automotive and construction sources.

Research is increasingly being directed to the recycling problems of waste glass from various industries and from domestic sources. Recycling of waste glass for many industries necessitates purification of the glass for its subsequent re-use. Nevertheless, due to the costs of purification, much of the waste glass is still committed to undesirable landfill sites. New applications for the use of waste glass would alleviate much of the waste disposal problems associated with waste glass.

According to a first aspect of the present invention, there is provided a solid glass composite matrix comprising glass granules and a binder resin which has set to bind the granules into a solid composite.

Preferably, the glass granules comprise between 40% and 90% w/w of the composite matrix, more preferably, between 65% and 85% w/w of the composite matrix, most preferably, between 75% and 85% w/w of the composite matrix.

When lower levels of glass granules are utilised other bulking sources may be added to the resin to top up the glass level. For instance, sand may be added to the mix as a bulking agent and to increase silica levels. As much as 50% bulking agent may be used, more appropriately as

preferably, at least 70% w/w of the glass composite matrix comprises glass granules of grain size 0mm-6mm, most preferably, at least 80% w/w of the glass composite matrix comprises glass granules of grain size between 0mm-6mm.

Preferably, at least 10% w/w of the glass composite matrix comprises glass granules of grain size 0mm-4mm, more preferably, at least 20% w/w of the glass composite matrix comprises granules of grain size 0mm-4mm, most preferably, at least 30% w/w of the glass composite matrix comprises granules of grain size 0-4mm.

Preferably, at least 10% w/w of the glass composite matrix comprises glass granules of grain size, 4mm-6mm, more preferably, at least 20% w/w of the glass composite matrix comprises glass granules of grain size 4mm-6mm, most preferably, at least 30% w/w of the glass composite matrix comprises glass granules of grain size 4-6mm.

Glass granules of between 6-10mm may also be present in the composite matrix. Granules between 6-10mm may be present at a level less than 50% w/w, more preferably, at a level less than 30% w/w, most preferably, at a level less than 25%.

Nevertheless, in some applications it is envisaged that 4-6mm granules or 6-10mm granules are present at up to 90% w/w, more preferably, up to 80% w/w, most preferably, up to 70% w/w of the composite matrix.

The waste glass may be derived from any suitable source including automotive, construction and consumer sources.

The coupling agent may be a silane coupling agent, preferably, an organo-functional silane coupling agent.

Preferably, the coupling agent is selected from a suitable silane, titanate ester or zirco-aluminate.

The resin may be selected from any suitable binder resin including epoxy resins, polyurethane binders, unsaturated polyester binders and poly C_1-C_2 alkyl methacrylate binders. Preferably, the polyalkyl methacrylate binder is polymethyl methacrylate.

A typical epoxy resin binder consists of the diglycidyl ether of bisphenol F or bisphenol A or mixtures thereof. Typically, the average number molecular weight is less than or equal to 1000, more preferably 800, most preferably, 700. A reactive diluent may be added to suit viscosity requirements. Typically, the reactive diluents comprise mono-functional or di-functional aliphatic or cycloaliphatic glycidyl ethers or esters. One or more of these may be mixed together in any proportions or used solely. A preferred diluent is a less viscous glycidyl ether such as C_{12}-C_{14} alkyl glycidyl ether. The specific diluent may be varied to suit viscosity requirements. Typically, the diluent is present at a level of 5-30% of the pre-cured resin, more preferably 10-25%.

The coupling agent may be present in the pre-cured resin at a level of 0.1-4.0% w/w, more preferably 0.5-3.0%, most preferably 1.0-2.0% w/w.

Advantages of the use of resin together with waste glass granules include the low level of chemical reactivity between the resin and the silica in the glass so that the composite produced is highly stable. Furthermore, it has been found that it is possible to introduce higher levels of glass in a resin substrate than alternative substrates. Due to contamination risks, preferably, the resin is substantially solvent free.

The composite of the invention provides an impervious surface which may be UV stable and has excellent chemical resistance against typical materials such as:

oil, petrol, diesel, anti-freeze, salts, beverages, urine and dilute acids and alkalis.

Advantageously, prior to setting, the composite may be shaped in three dimensions and inconsistencies in the final set shape may be simply corrected by filling or polishing as is necessary. The casting techniques may be any of those known to those skilled in the art including vacuum-, pressure- and vibro-casting.

The composites of the invention may be utilised in many applications including:

internal and external flooring, furniture, lighting, work surfaces, architectural features such as skirting, architraves and sanitary work and the invention extends to methods of making such products using the method of the second aspect of the invention or the product of the first aspect. Furthermore, impervious examples of the product

percentages and absolute weights of the various constituents of the composites.

The epoxy resin in examples 1-10 comprises a blend of 80-84% bisphenol A & F, 15-19% C_{12}-C_{14} alkyl glycidyl ether as a diluent and 1% glycidoxy-functional silane coupling agent.

All these products produced high quality products after grinding and polishing.

The method of preparation of composite is as follows.

i. The glass is weighed out in the correct percentages of each grain size and colour. (Usually 0-4mm, 4-6mm and 6-10mm, in colours green bottle, amber bottle, blue bottle and clear plate). See examples.

ii. The resin binder is mixed to the correct ratios (Base, catalyst and pigment), see examples, until an even dispersion is achieved.

iii. The glass is then mixed thoroughly into the mixed resin binder.

iv. The "mix" is then either trowelled into moulds (for production of tiles or three dimensional items eg. furniture) or laid directly onto a preprepared floor surface as a screed.

Curing times @ 20°C

24 hours ... 70%

7 days ... 95%

28 days ... 100%

Although all curing times can be varied by using additives.

Table 2 (Example 2)

Constituency	Percentage	Weight	Comments
Epoxy resin	11.67	262.92	Clear
Octahydro-4,7-methano-1H-indendimethyl amine,	5.89	132.75	Clear
Pigment RAL No. 6019 Bs No.	0.12	2.60	
Aggregate mm 0.4	40.87	920.50	Clear Plate
Aggregate mm 4-6	41.45	933.75	Clear Plate
Aggregate mm 6-10	0	0	
Total	100	2252.52	

Table 4 (a) (Example 4a)

Constituency	Percentage	Weight	Comments
Epoxy resin	10.95	93.68	Clear
Octahydro-4,7-methano-1H-indendimethyl amine.,	5.27	45.05	Clear
Pigment RAL No. 9003	0.32	2.73	
Aggregates 0.4mm	58.13	497.25	Blue
4-6mm	25.34	216.75	Blue
6-10mm	0.00	0.00	
Total	100	855.46	

Table 5

Constituency	Percentage	Weight	Comments
Epoxy resin	14.04	120.15	Clear
Octahydro-4,7-methano-1H-indendimethyl amine.,	6.85	58.65	Clear
Pigment RAL No.9003 5005 BS No.	 0.04 0.01	 0.30 0.10	
Aggregates 0-4mm	 19.87	 170.00	 Clear Plate
4-6mm	38.74 0.58	331.50 5.00	Clear plate Green
6-10mm	19.87	170.00	
Total	100.00	855.70	

Tables 6-10 reveal grain size distribution for suitable glass granular samples which may be used with resins in accordance with the present invention.

Table 8

Grain Size (mm)	Mass (g)	Percentage
> 4	78.60	7.86
3.15-4	127.00	12.70
2-3.15	239.00	23.90
1-2	244.00	24.40
0.71-1	114.50	11.45
0.5-0.71	75.10	7.51
0.25-0.5	91.30	9.13
0-0.25	30.50	3.05

Table 9

Grain size (mm)	Mass (g)	Percentage
> 4	134.03	13.40
3.15-4	192.06	19.21
2-3.15	339.50	33.95
1-2	174.20	17.42
0.71-1	52.58	5.26
0.5-0.71	31.28	3.13
0.25-0.5	44.60	4.46
0-0.25	31.75	3.17

Table 11

	Recorded Penetration
Test No 1	0.5mm
Test No 2	0.4mm
Test No 3	0.2mm
Test No 4	0.0mm

Table 12

	Mean Coefficient of Thermal Expansion
Test age 7 days	3.0×10^{-5}
Test age 28 days	4.2×10^{-5}

Table 13 shows that the flexural strength of example 8 is very high and this may give opportunities in combination with flexible backing surfaces or substrates such as soil or earth.

Table 13

	Flexural Strength (N/mm^2)
Test age 7 days	(See Note 1)
Test age 28 days	41.9 (see Note 2)

Note 1 - Test pieces flexed without failure, flexural strength can not be recorded.

Note 2 - Test pieces 1 and 3 had air bubbles on the fracture surface, test 2 flexed a long way before failure.

Table 16 (Example 6)

Impact Testing (Sample)

Constituency	Percentage	Weight	Comments
Epoxy resin	11.17	251.67	Clear
Octahydro-4,7-methano-1H-indendimethyl amine.	5.65	127.35	Clear
Pigment RAL No. 2010 BS No.	0.06	1.25	
Aggregates 0-4mm	42.66	960.97	Clear Plate
4-6mm	40.46	911.25	Clear Plate
6-10mm	0.00	0.00	
TOTAL	100.00	2252.49	

Table 18 (Example 8)

Flexural Strength Sample

Constituency	Percentage	Weight	Comments
Epoxy resin	11.17	251.67	Clear
Octahydro-4,7-methano-1H-indendimethyl amine.	5.65	127.35	Clear
Pigment RAL No. 2010	0.06	1.25	
Aggregates 0-4mm	42.66	960.97	Clear Plate
4-6mm	40.46	911.25	Clear Plate
6-10mm	0.00	0.00	
TOTAL	100.00	2252.49	

Table 20 (Example 10)
Coefficient of Friction Sample

Constituency	Percentage	Weight	Comments
Epoxy resin	11.17	251.67	Clear
Octahydro-4,7-methano—1H-indendimethyl amine.,	5.65	127.35	Clear
Pigment RAL 2010	0.06	1.25	
Aggregates 0-4mm	42.66	960.97	Clear Plate
4-6mm	40.46	911.25	Clear Plate
6-10mm	0	0	
TOTAL	100.00	2252.49	

The reader's attention is directed to all papers and documents which are filed concurrently with or previous to this specification in connection with this application and which are open to public inspection with this specification, and the contents of all such papers and documents are incorporated herein by reference.

All of the features disclosed in this specification (including any accompanying claims, abstract and drawings), and/or all of the steps of any method or process so disclosed, may be combined in any combination, except combinations where at least some of such features and/or steps are mutually exclusive.